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International application number: PCT/EP05/051404

International filing date: 27 March 2005 (27.03.2005)

Document type: Certified copy of priority document

Document details: Country/Office: FR
Number: 0403905
Filing date: 13 April 2004 (13.04.2004)

Date of receipt at the International Bureau: 14 October 2005 (14.10.2005)

Remark: Priority document submitted or transmitted to the International Bureau in compliance with Rule 17.1(a) or (b)

PCT/EP200 5 / 0 5 1 4 0 4

R E P U B L I Q U E  F R A N Ç A I S E

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# BREVET D'INVENTION

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Le Chef du Département des brevets

Martine PLANCHE

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SIEGE
26 bis, rue de Saint-Petersbourg
75800 PARIS cedex 08
Téléphone : 33 (0)1 53 04 53 04
Télécopie : 33 (0)1 53 04 45 23
www.inpi.fr

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The Head of the Patent Department

[signed]

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**1** NOM ET ADRESSE DU DEMANDEUR OU DU MANDATAIRE À QUI LA CORRESPONDANCE DOIT ÊTRE ADRESSÉE

DESBRANDES Robert
Allée des Chériniers
03190 GIVARLAIS

**Vos références pour ce dossier** *(facultatif)* DVG_RD_#1

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**3 TITRE DE L'INVENTION** (200 caractères ou espaces maximum)

Procédé et Appareillage pour modifier la probabilité de désexcitation des nucléides isomères.

**4 DÉCLARATION DE PRIORITÉ OU REQUÊTE DU BÉNÉFICE DE LA DATE DE DÉPÔT D'UNE DEMANDE ANTÉRIEURE FRANÇAISE**

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| Prénoms | | Robert |
| Forme juridique | | |
| N° SIREN | | |
| Code APE-NAF | | |
| Domicile ou siège | Rue | Allée des Chériniers |
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| Adresse électronique *(facultatif)* | | rdesbrandes@wanadoo.fr |

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| **9 REDUCTION OF THE FEES**<br>**(refer to Explanatory leaflet Heading 9)** | |
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| **10 SEQUENCES OF NUCLEOTIDES AND/OR OF AMINO ACIDS** | |
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# PATENT
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The present invention relates to a method and an equipment to modify the probability of deexcitation of the isomer nuclides.

The probability of deexcitation of a radioactive body is connected to the half-life, i.e. time necessary to the deexcitation of half of the radioactive nuclei. This probability is given the formula:

$$P = LN(2) / \lambda$$

P, probability of deexcitation per minutes;

LN, natural logarithm;

$\lambda$, half-life in minutes.

For example, the half-life of normal indium $115^{m}$ is 268 minutes. The probability of deexcitation of a nucleus per minutes is thus 0,00258 that represents one chance on 387 per minute. By normal indium $115^{m}$, one indicates classically excited isomer and not as stipulated in this invention.

There are many nuclides, which have a metastable state (isomers) whose half-life goes, according to isomers, from one second or less to 50 years or more. A list of the main isomers is given in table 1. In this table are listed the symbol, the abundance of the isotope, the half-life of the nuclei and the energy of the gamma radiation emitted at the time of the deexcitation. Indium 115 for example has a 268 minutes metastable state (4.48 hours) of half-life as shown in the figure 1. It returns to its stable fundamental state by isomeric transition while emitting a gamma radiation from 336.2 keV. The isomeric transition, like internal conversion, does not change the atomic number. In its normal state, an isomer returns to its fundamental state with the half-life mentioned in table 1. Certain isomer nuclei, like Hafnium 178 or Hafnium 179, emit several gamma at the time of their return to the fundamental state.

It is known to the experts that the deexcitation of

the isomer can be accelerated by X or gamma irradiation. In this invention the half-life of the isomer changes with time without the intervention of an irradiation, sometimes called X or gamma stimulation. In addition, the half-life obtained within the framework of the invention, varies with the time while being shorter at the beginning of the life of the isomer, and longer thereafter as shown in figure 5 in the case of indium $115^{m}$.

The radioactive elements have a rigorously constant half-life within the limits of the statistical fluctuations. Except for the case of isomers irradiated according to the methods described in the present invention or the stimulation, it is impossible to vary the half-life of a radioactive isomer. This invention thus solves a technical problem by providing a radioactive element with a half-life variable without stimulation and adaptable for a given application.

The probability of disintegration or deexcitation of a radioactive element is not modified by a change of its physical or chemical state. Consequently, the excited samples with the techniques described in this invention can be transformed by fusion, vaporization, dissolution or chemical combination after irradiation without modification of their nuclear properties.

Several isotopes can exist naturally or be artificially incorporated in the samples. These samples can then be alloys or mixtures of several isotopes having a metastable state. In this case, the half-life of each excited isotope according to the invention can be measured simultaneously with a gamma spectrograph, which is known of the expert.

Various industrial or medical applications are possible. A chemical reaction for example can require a strong dose of radiation at the beginning, that is followed by a weaker dose and lasting a long time. It is the same

for a medical treatment, which requires an evolution of the doses in time. The use of several isotopes in the same sample is used to have simultaneously gamma of various energies at the time of the natural deexcitation of the isotopes.

The invention, whose implementation will be detailed in the continuation, is not explained by the admitted nuclear theories at present. Consequently, it does not result from a known technique of the expert.

The method according to the invention consists in irradiating, using gamma rays, a sample of an element or several elements having a metastable state with a half-life duration going from less than one second to several years. The radiation source can be either a radioactive isotope, or a linear accelerator of particles, such as electrons, alpha or protons particles, which by Bremstrahlung effect produce gamma rays.

In the case of the radioactive source, the gamma rays must be emitted in a cascade by the same nucleus. For example, an emission in a cascade is provided by cobalt 60, as shown in figure 2. The emitted gamma rays must have a sufficient energy to carry out a reverse isomeric transition, i.e. to have the nucleus go from its fundamental state to its metastable state. In the case of indium 115, for example, the necessary energy is 1080 keV, this condition is met by the two gamma rays of cobalt 60. One sees on figure 2 that one of the gamma has an energy of 1173 keV with 99.90% chances to occur, and the other 1332 keV with 99.98% chances to occur. A cascade thus occurs, because two gamma are emitted with an interval of 0.713 picosecond ($10^{-12}$ S) as an average.

In the case of an irradiation by the Bremstrahlung gamma rays of a linear accelerator of particles, for example of electrons, the energy of gamma must again be higher than the threshold of excitation of the selected

element.

For example, a compact linear accelerator can issue a gamma radiation very focused with a spectrum of gamma energies from 0 to 6 MeV. This spectrum is reproduced in figure 3. The energy of all the electrons before striking the tungsten target is 6 MeV. Consequently, each electron emits on average four gamma of 1.5 MeV (1500 keV) as shown in figure 3 in a very fast succession comparable to a cascade. The cascade of gamma obtained with the compact linear accelerator is, as the experiment shows it, more effecient to modify the half-life than the source of cobalt 60.

According to a particular mode of the invention, the samples to be irradiated are placed on a tray (3), which presents the samples (5) in succession in front of a piston (7) which introduces them opposite a radioactive source (1) by the opening (4) as shown in the figure 4. The source is placed in a thick steel and lead shielding (2). An axis (6) connects the tray to a motor (10) controlled by a timer (11). The time of irradiation is adjusted for each sample using a timer (9), which actuates a pneumatic valve (8) to obtain the optimal response of activation.

In the case, for example, of indium 115, a 20 hours irradiation with a source of 111000 GBq (3000 Ci) of cobalt 60 produces the indium isomer with a 242 minutes initial half-life instead of 268 minutes, which is the half-life of the normal isomer, which is a reduction of 10%. This reduction can be modified by varying the time of irradiation. Contrary to the normal isomer, as from 1500 minutes of elapsed time, the half-life exceeds the normal half-life of 268 minutes to reach 360 minutes after 3000 minutes elapsed. The sample thus remains slightly radioactive for a very long time. Figure 5 schematically shows the evolution of the half-life for an indium sample irradiated ($In\ 115^{m}$) under the preceding conditions.

According to another mode of implementation of the invention, schematized on figure 6, the samples (14) are placed on a rotary tray (13). This tray is supported by an axis (15) and is connected to a motor (16), such motor being controlled by a timer (17). The samples are presented in sequence in front of the beam of x-rays of a compact linear accelerator (12) for example. A "phantom" (18) filled with water stops the not absorbed gamma rays. In general the accelerators cannot function permanently. A certain number of units of time of irradiation, for example 5 minutes, is applied to each sample according to the desired initial half-life using a timer (19).

The accelerator emits a focused radiation, contrary to a source of cobalt 60. Moreover, in the preceding example, up to four gamma with a sufficient energy to activate nuclei, such as the indium 115 nuclei, are produced in a cascade. This radiation is thus more effecient and a short time of irradiation is generally sufficient. Figure 7 represents the schematic evolution of the half-life of an indium 115 sample irradiated with a compact linear accelerator during 20 minutes. The initial half-life is 130 minutes as compared to 268 minutes for normal $In^{115m}$, that is to say a reduction of 50%. Again the normal half-life is reached after 1500 minutes elapsed and then increases to a half-life of 400 minutes when the time reaches 3000 minutes elapsed.

The equipment described previously is examples of implementation. Other means to present the samples at the irradiation can be employed without leaving the framework of the invention.

The samples to be irradiated are solids in sheets or powder, fluids or gases (case of Xenon for example), which contain a proportion of one or several isotopes of table 1. The samples can be also alloys, mixtures or chemical compounds incorporating a proportion of one or several

isotopes of table 1. The samples can also be transformed physically or chemically after irradiation. For example, a sample in the form of powder or of a gas can be incorporated in injectable carrying molecules.

The measures of half-life can be taken with the conventional instruments of the expert. The gamma spectroscopes used at present contain thousands of channels to simultaneously measure the response of hundreds of radioactive or excited isotopes.

A common instrument is a germanium crystals detector functioning at low temperature. In order to minimize the effects of the cosmic rays, of radon and of the ambient interferences, the samples are placed in a container with walls of copper, lead, and steel. An analyzer is set on one or several characteristic radiations of one or several selected isomers. For example, in the case of indium $115^{m}$, gamma in the 336 keV line are counted. In the case of Hafnium 179, having a 25 days of half-life, many lines are detectable whose main ones are 453, 409, 362, 315, 268, and 122 keV. These lines are emitted in a cascade with picoseconds of interval and are easily detected by the spectrographs with germanium crystals. It is also possible that progress of the technique will make it possible to measure the radiation of 336 keV without a special container.

# CLAIMS

1) Method and equipment to modify the probability of deexcitation, therefore the half-life, of the isomer nuclides, characterized by the irradiation of a sample containing an isotope having a metastable state by a source of gamma rays emitted in a cascade, either by a radioactive source, or by a generator of gamma rays coming from Bremstrahlung of accelerated particles, with a sufficient energy to excite the aforementioned element to its metastable state, and a sufficient duration to obtain the necessary initial half-life.

2) Method and equipment according to claim 1 characterized by the use of a sample containing a plurality of isotopes having a metastable state of a half-life from 1 second to 50 years.

3) Method and equipment according to claim 2 characterized by the use of gamma rays issued in a cascade and an energy higher than the threshold of excitation of the isotopes used that have a metastable state.

4) Method and equipment according to claim 2 characterized by the use of samples containing several isotopes of which the gamma emission of each one of them is measured simultaneously.

5) Method and equipment according to claim 2 characterized by the use of samples containing several isotopes of which the gamma emission is made up of a plurality of lines measured simultaneously.

6) Method and equipment according to claim 2 characterized by the use of samples in various physical forms.

CLAIMS

1) A method for modifying the probability of deexcitation, therefore the half-life, of the isomer nuclides, in which:

- one prepares a sample containing at least an isomer nuclide having a metastable state by irradiation with the means either of a source of gamma rays emitted in a cascade, or of a generator of gamma rays coming from Bremstrahlung of accelerated particles, with an energy higher than the threshold of excitation of the aforesaid isomer nuclide to excite the aforementioned isomer nuclide in his metastable state,

characterized:

- in that the initial half-life of each excited isomer nuclide of the sample obtained previously is lower than the theoretical half-life of the aforesaid nuclide, mentioned in the tables of isotopes, and that these initial half-lives vary with the time elapsed and the power of the irradiation source,

- in that one uses the gamma emission of variable instantaneous half-life from at least one excited isomer nuclide, during its natural deexcitation, and in that the value of the half-life of the aforesaid nuclide varies from the value of the initial half-life to the theoretical half-life of the aforesaid nuclide, then increases beyond this value of the aforesaid theoretical half-life.

2) Method according to claim 1 characterized in that one uses samples containing at least an isomer nuclide having a metastable state, for example: Niobium (93Nb41m), Cadmium (111Cd48m), Cadmium (113Cd48m), Cesium (135Ce55m),

7) Method and equipment according to claim 2 characterized by the use of samples in various chemical forms.

8) Method and equipment according to claim 2 characterized by the use of a sample in the form of a solution.

9) Method and equipment according to claim 2 characterized by the use of a sample having undergone a physical transformation after irradiation.

10) Method and equipment according to claim 2 characterized by the use of a sample having undergone a chemical transformation after irradiation.

Indium (115In49m), Tin (117Sn50m), Tin (119Sn50m), Tellurium (125Te52m), Xenon (129Xe54m), Xenon (131Xe54m), Hafnium (178Hf72m), Hafnium (179Hf72m), Iridium (193Ir77m), Platinum (195Pt78m).

3) Method according to anyone of the claims 1 or 2 characterized in that one uses samples containing several excited isomer nuclides of which the gamma emission of each one of them is measured simultaneously.

4) Method according to anyone of the claims 1, 2 or 3 characterized in that one uses samples containing at least an excited isomer nuclide of which the gamma emission is made up of a plurality of lines measured simultaneously.

5) Method according to anyone of the claims 1, 2, 3 or 4 characterized in that the measured initial half-life value of at least one isomer nuclide is comprised between 10% and 100% of the theoretical value.

6) Method according to anyone of the claims 1, 2, 3, 4 or 5 characterized in that one uses samples in various physical forms or various chemical forms.

7) Method according to anyone of the claims 1, 2, 3, 4, 5 or 6 characterized in that one uses a sample in the form of a solution.

8) Method according to anyone of the claims 1, 2, 3, 4, 5, 6 or 7 characterized in that one uses a sample having undergone a physical transformation or a chemical transformation after irradiation.

9) Device to implement the method according to anyone of the claims 1 to 8 characterized in that it comprises:

- An equipment of excitation that irradiates a sample containing at least an isomer nuclide having a metastable state with the means either of a source of

gamma rays emitted in a cascade, or of a generator of gamma rays coming from Bremstrahlung of accelerated particles, with an energy higher than the threshold of excitation of the aforesaid isomer nuclide to excite it to its metastable state,

- An equipment controlling the time of the irradiation of each sample according to the required half-life.

10) Use of the method according to anyone of the claims 1 to 8 to provide a low dose of radiations for a long time, starting from an initial high dose of radiations.

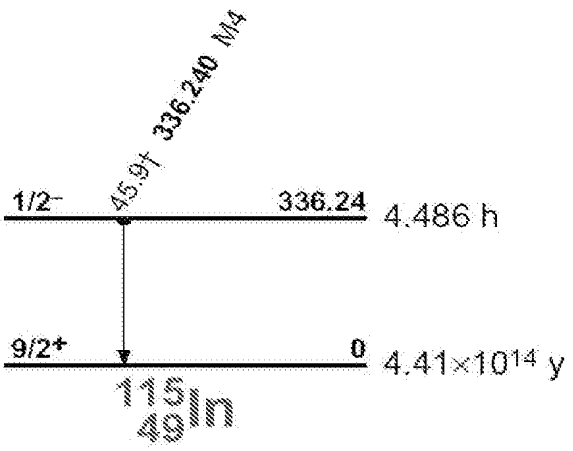

336.240 M4
45.9†
1/2⁻
336.24
4.486 h
9/2⁺
0
4.41×10¹⁴ y
115 49In

**Fig. 1**

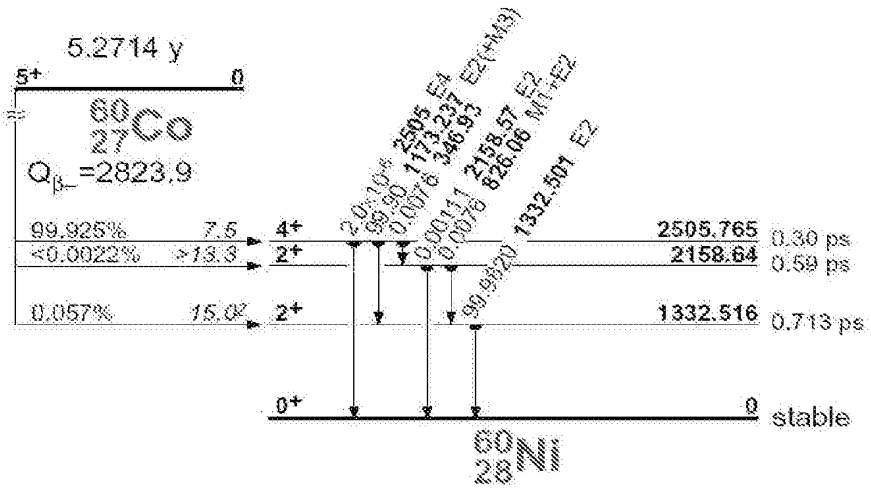

5.2714 y
5⁺
0
60 27Co
Qβ⁻=2823.9
99.925%
7.5
<0.0022%
>13.3
0.057%
15.0?
4⁺
2⁺
2⁺
0⁺
0
2505.765
2158.64
1332.516
0.30 ps
0.59 ps
0.713 ps
stable
2.0×10⁻⁶
2505 E4
99.88
1173.237 E2(+M3)
0.0076
346.93
0.00111
2158.57 E2
0.0076
626.06 M1+E2
99.9880
1332.501 E2
60 28Ni

**Fig. 2**

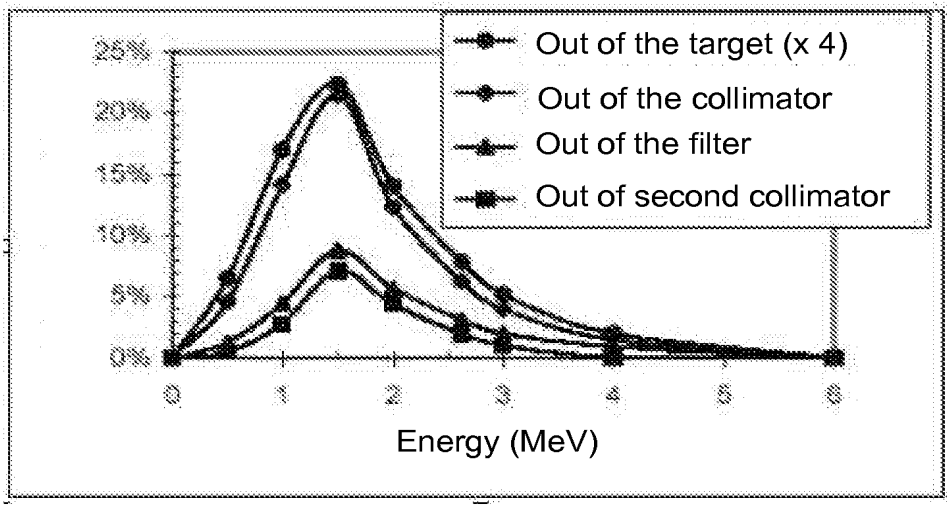

Out of the target (x 4)
Out of the collimator
Out of the filter
Out of second collimator
25%
20%
15%
10%
5%
0%
0
1
2
3
4
5
6
Energy (MeV)

**Fig. 3**

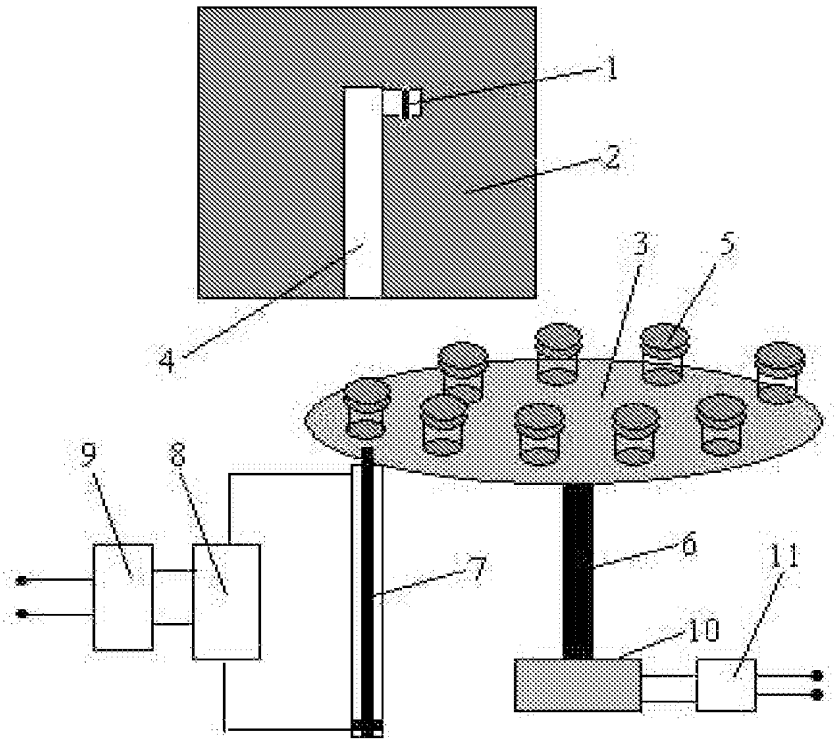

1
2
3
4
5
6
7
8
9
10
11

**Fig. 4**

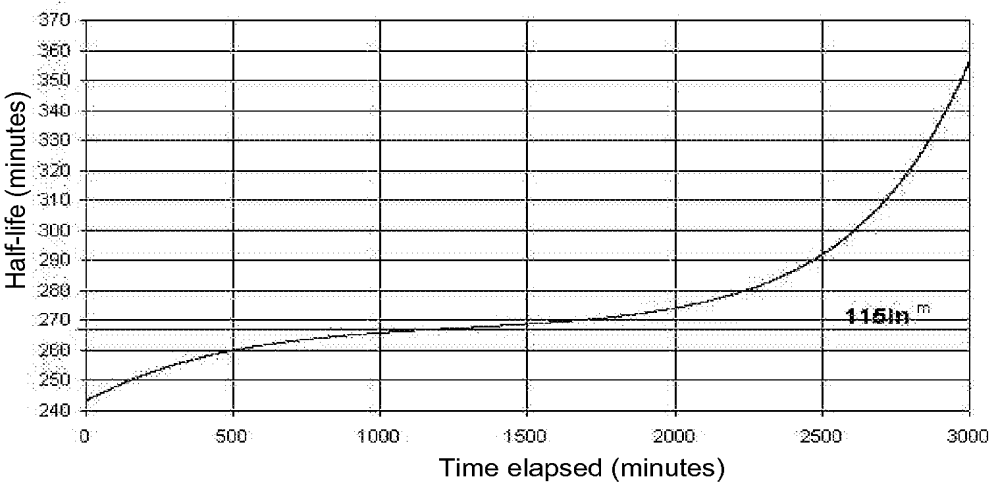

Half-life (minutes)
370
360
350
340
330
320
310
300
290
280
270
260
250
240
0
500
1000
1500
2000
2500
3000
Time elapsed (minutes)
115In m

**Fig. 5**

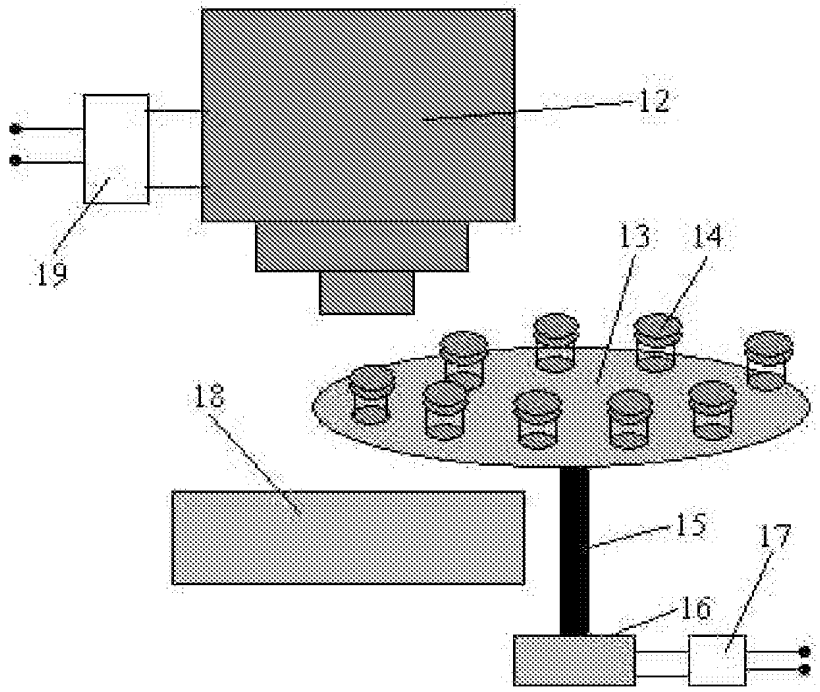

12
19
13
14
18
15
17
16

**Fig. 6**

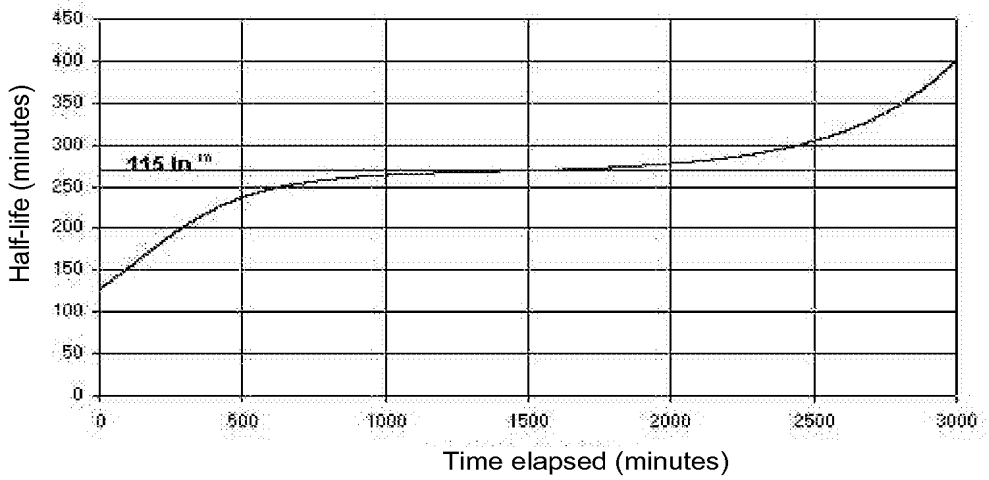


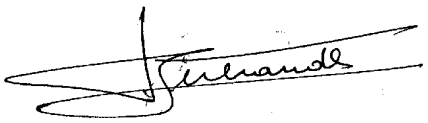

**Fig. 7**

| Nuclide | Symbol | Abundance % | Half-life | Gamma keV |
|---|---|---|---|---|
| Niobium | 93Nb41 | 100 | 16.3 y | 31.8 |
| Cadmium | 111Cd48 | 12.8 | 48.54 m | 396.2 |
| Cadmium | 113Cd48 | 12.2 | 14.1 y | 263.5 |
| Cesium | 135Ce | - | 53 m | 846/786 |
| Indium | 115In49 | 95.7 | 4.48 h | 336.2 |
| Tin | 117Sn50 | 7.7 | 13.6 y | 314.6 |
| Tin | 119Sn50 | 8.6 | 293 d | 60.5 |
| Tellurium | 125Te52 | 7.1 | 57.4 d | 144.8 |
| Xenon | 129Xe54 | 26.5 | 8.8 d | 238.1 |
| Xenon | 131Xe54 | 21.2 | 11.8 d | 163.9 |
| Hafnium | 178Hf72 | 27.3 | 31 y | 574/…./93 |
| Hafnium | 179Hf72 | 13.6 | 25 d | 453/…./122 |
| Iridium | 193Ir77 | 62.7 | 10.5 d | 80.2 |
| Platinum | 195Pt78 | 33.8 | 4 d | 259.3 |

m: minutes, h: hours, d: days, y: years.

**Table 1**

I certify that this document is the English translation, to the best of my knowledge, of the document referenced in the first page.



Robert DESBRANDES, inventor